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Relaxation of nuclear spin in atomic hydrogen due to long-range orbital currents in metal walls

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Thermal current fluctuations in bulk metal produce a fluctuating magnetic field that extends outside the metal's surface. We find that its spectral density $\langle H(z)^2 \rangle_\omega$ as a function of distance from the surface decays as $\ln(z)$ at distances less than the mean free path of the electrons in the metal and as $1/z$ at large z. We evaluate the nuclear-spin relaxation rate due to these fluctuations. We apply our model to the one-body surface relaxation rate of atomic hydrogen. We find the relaxation time to be of the order of 10^3 s at a temperature of 1 K. A possible relaxation mechanism due to electric-field fluctuations, which may also be applicable to cells with insulating walls, is discussed.

I. INTRODUCTION

The quest to observe Bose-Einstein condensation has led to the study of the relaxation and recombination processes in spin-polarized atomic hydrogen. These processes at modest densities take place mainly on the surface of the cell containing the gas. The decay of atomic hydrogen was found to be dominated by some one-body relaxation process on the surface.[1-5] This relaxation is essentially a transition between the two lowest hyperfine levels, which is equivalent at high magnetic fields to the nuclear-magnetic-moment transition between Zeeman levels. The one-body surface relaxation rate observed in the above experiments was unexpectedly high. Berlinsky, Hardy, and Statt[3,6] have attempted to explain this phenomenon in terms of the interaction of the spin with magnetic impurities in the substrate. This explanation was initially supported by achievement of slower-relaxation rates through copper purification. But further purification did not affect the relaxation time suggesting the existence of another mechanism. In this paper we suggest a mechanism that invokes magnetic-field fluctuations produced by thermal current fluctuations in the metallic cell walls.

We note that there are two types of experiments. They differ by the kind of materials that the cells containing hydrogen are made of. The first type involves cells with copper walls and our mechanism should be applicable to these experiments. The second type[5] involves quartz walls, to which our explanation is not applicable. The observation of the high relaxation rate in this case suggests the existence of some other mechanism. We discuss one possible mechanism in the Appendix.

Our idea can be illustrated by the following simple consideration. Suppose the half space defined by $z<0$ is filled with metal at temperature T. We are interested in the spectral density of the mean square value of the magnetic field, $\langle H^2 \rangle = \int_{-\infty}^{+\infty} dt\, e^{-i\omega t} \langle H(\mathbf{r},0) H(\mathbf{r},t) \rangle$, at a distance z from the surface ($z>0$) in the limit $\omega \rightarrow 0$. Let us divide the metal into layers of thickness Δr parallel to the surface, and divide each layer into a series of concentric annuli with width Δa and centered on the z axis. We will calculate contributions to the magnetic field from each loop and then sum them. The magnetic field from one loop of radius a, carrying current ΔI, at distance r from its center is given by $\Delta H \sim a^2 \Delta I / (a^2 + r^2)^{3/2}$. The spectral density of the mean square current in one loop can be expressed as $\langle \Delta I^2 \rangle = T / \Delta R$, where ΔR is the resistance. We can write $(\Delta R)^{-1} = \sigma \Delta a \Delta r / 2\pi a$ where σ is the conductivity of the metal. Thus we obtain $\langle \Delta H^2 \rangle \sim T \sigma a^3 \Delta a \Delta r / (a^2 + r^2)^3$. Now we can sum all the contributions by integrating this expression with respect to a from 0 to ∞ and with respect to r from z to ∞; this gives $\langle H(z)^2 \rangle \sim T\sigma / z$. Therefore one might expect a slow decay of the magnetic-field correlation with growing distance from the metal surface.

In this paper we carry out a detailed calculation of the relaxation rate of a nuclear spin with magnetic moment μ in an external magnetic field H_0 due to the mechanism described above. The spin is located a distance z from the metal surface, which is at temperature T. To do this we need the magnetic-field correlation function, $\langle |H^\alpha(z,\omega)|^2 \rangle$, where α is a spatial index and ω is the frequency of the transition ($\hbar\omega = 2\mu H_0$). We relate the magnetic field to conduction currents using Maxwell's equations and neglect the displacement current term, assuming that $\sigma \gg \omega$ and $z \ll c/\omega$. Our work extends an earlier calculation done by Lee and Nagaosa[7] for the case of a spin located between parallel metallic layers. That earlier work was done in the context of high-T_c superconductors, whereas here we are more concerned with conventional metals.

The transverse current-current correlation function $\langle j^\alpha(\mathbf{k}) j^\beta(\mathbf{k}') \rangle$ is obtained from the transverse conductivity $\sigma_\perp(\mathbf{k})$. Then we give an estimate of the relaxation rate for atomic hydrogen atoms adsorbed on a liquid helium layer so that the atoms are at a constant distance from the metal surface. Comparison with experiments will be discussed.

II. CALCULATION OF THE RELAXATION RATE

The relaxation rate in the external field H_0 in the z direction is given by[8]

$$\frac{1}{T_z}=\frac{\gamma^2}{2}[\langle|H^y(\mathbf{r},\omega)|^2\rangle+\langle|H^x(\mathbf{r},\omega)|^2] , \tag{1}$$

where $\gamma=\mu/\hbar I_z$ (I_z is the spin component in the z direction). In case of the proton this gives $\gamma=2\mu/\hbar$. We define the Fourier transformation in the following way:

$$\langle|H^\alpha(\mathbf{r},\omega)|^2\rangle =\frac{1}{(2\pi)^6}\int_{-\infty}^{+\infty}d\mathbf{p}\,d\mathbf{p}'e^{i(\mathbf{p}+\mathbf{p}')\cdot\mathbf{r}}\langle H^\alpha(\mathbf{p})H^\alpha(\mathbf{p}')\rangle_\omega , \tag{2}$$

$$H^\alpha(t)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}d\omega\, e^{i\omega t}H^\alpha(\omega) . \tag{3}$$

Index ω in the expressions of the type $\langle H^\alpha(\mathbf{p})H^\alpha(\mathbf{p}')\rangle_\omega$ will be omitted in the rest of the paper. Maxwell's equations yield, under Fourier transformation,

$$H^x(\mathbf{p})=\frac{4\pi i}{c}\frac{[p_y j^z(\mathbf{p})-p_z j^y(\mathbf{p})]}{p^2} , \tag{4}$$

$$H^y(\mathbf{p})=\frac{4\pi i}{c}\frac{[p_z j^x(\mathbf{p})-p_x j^z(\mathbf{p})]}{p^2} \tag{5}$$

$$H^z(\mathbf{p})=\frac{4\pi i}{c}\frac{[p_x j^y(\mathbf{p})-p_y j^x(\mathbf{p})]}{p^2} \tag{6}$$

These relations lead to the following expressions for $\langle H^\alpha(\mathbf{p})H^\alpha(\mathbf{p}')\rangle$:

$$\langle H^x(\mathbf{p})H^x(\mathbf{p}')\rangle=-\frac{(4\pi)^2}{c^2p^2p'^2}[p_zp_z'\langle j^y(\mathbf{p})j^y(\mathbf{p}')\rangle+p_yp_y'\langle j^z(\mathbf{p})j^z(\mathbf{p}')\rangle-p_zp_y'\langle j^y(\mathbf{p})j^z(\mathbf{p}')\rangle -p_yp_z'\langle j^z(\mathbf{p})j^y(\mathbf{p}')\rangle] , \tag{7}$$

$$\langle H^y(\mathbf{p})H^y(\mathbf{p}')\rangle=-\frac{(4\pi)^2}{c^2p^2p'^2}[p_xp_x'\langle j^z(\mathbf{p})j^z(\mathbf{p}')\rangle+p_zp_x'\langle j^x(\mathbf{p})j^x(\mathbf{p}')\rangle-p_xp_z'\langle j^z(\mathbf{p})j^x(\mathbf{p}')\rangle -p_zp_x'\langle j^x(\mathbf{p})j^z(\mathbf{p}')\rangle] , \tag{8}$$

$$\langle H^z(\mathbf{p})H^z(\mathbf{p}')\rangle=-\frac{(4\pi)^2}{c^2p^2p'^2}[p_xp_x'\langle j^y(\mathbf{p})j^y(\mathbf{p}')\rangle+p_yp_y'\langle j^x(\mathbf{p})j^x(\mathbf{p}')\rangle-p_xp_y'\langle j^y(\mathbf{p})j^x(\mathbf{p}')\rangle -p_yp_x'\langle j^x(\mathbf{p})j^y(\mathbf{p}')\rangle] . \tag{9}$$

For a bulk metal which is translationally invariant, we expect

$$\langle j_0^\alpha(\mathbf{k})j_0^\beta(\mathbf{k}')\rangle=2T\sigma_{\alpha\beta}(\mathbf{k})(2\pi)^3\delta(\mathbf{k}+\mathbf{k}') , \tag{10}$$

where the subscript 0 designates the value for a bulk metal. However, we cannot use (10) directly in (7)–(9) because currents can exist only in the half space. To overcome this difficulty we calculate the Fourier transform of $\langle j_0^\alpha(\mathbf{k})j_0^\beta(\mathbf{k}')\rangle$ in coordinate space:

$$\langle j_0^\alpha(\mathbf{r})j_0^\beta(\mathbf{r}')\rangle =\frac{1}{(2\pi)^6}\int_{-\infty}^{+\infty}d\mathbf{k}\,d\mathbf{k}'e^{i(\mathbf{k}\cdot\mathbf{r}+\mathbf{k}'\cdot\mathbf{r}')}\langle j_0^\alpha(\mathbf{k})j_0^\beta(\mathbf{k}')\rangle . \tag{11}$$

A simple procedure would be to transform back to the momentum space, integrating only over $z<0$:

$$\langle j^\alpha(\mathbf{p})j^\beta(\mathbf{p}')\rangle =\int_{z,z'<0}d\mathbf{r}\,d\mathbf{r}'e^{-i\mathbf{p}\cdot\mathbf{r}-\mathbf{p}'\cdot\mathbf{r}'}\langle j_0^\alpha(x,y,z)j_0^\beta(x',y',z')\rangle . \tag{12}$$

However, this is not adequate because the newly obtained correlation functions $\langle j^\alpha(\mathbf{p})j^\beta(\mathbf{p}')\rangle$ violate current conservation; namely, using the equation $\mathbf{p}\cdot\mathbf{j}(\mathbf{p})=0$ we should be able to express one correlation function in terms of the others. It is easy to check that this is not the case for Eq. (12). For example, the equation

$$-p_y'\langle j^x(\mathbf{p})j^y(\mathbf{p}')\rangle=p_x'\langle j^x(\mathbf{p})j^x(\mathbf{p}')\rangle +p_z'\langle j^x(\mathbf{p})j^z(\mathbf{p}')\rangle \tag{13}$$

is not satisfied if correlation functions are given by (12). This happened because the assumption of replacing $\langle j^\alpha(\mathbf{r})j^\beta(\mathbf{r}')\rangle$ by the bulk value $\langle j_0^\alpha(\mathbf{r})j_0^\beta(\mathbf{r}')\rangle$ breaks down near the metal-vacuum interface. To avoid this inconsistency we introduce the following ansatz:

$$j^x(z)=\alpha[j_0^x(x,y,z)+j_0^x(x,y,-z)] , \tag{14}$$

$$j^y(z)=\alpha[j_0^y(x,y,z)+j_0^y(x,y,-z)] , \tag{15}$$

$$j^z(z)=\alpha[j_0^z(x,y,z)-j_0^z(x,y,-z)] , \tag{16}$$

which ensures that the perpendicular component of the current vanishes at the interface. Physically our ansatz means that the current undergoes specular reflection at the surface. Now we substitute this into the right-hand side of (12). For example, for $\langle j^y(\mathbf{p})j^y(\mathbf{p}')\rangle$, instead of (12) we have

$$\langle j^y(\mathbf{p})j^y(\mathbf{p}')\rangle=\tfrac{1}{2}\int_{z,z'<0}d\mathbf{r}\,d\mathbf{r}'e^{-i\mathbf{p}\cdot\mathbf{r}-\mathbf{p}'\cdot\mathbf{r}'}[\langle j_0^y(x,y,z)j_0^y(x',y',z')\rangle+\langle j_0^y(x,y,-z)j_0^y(x',y',-z')\rangle$$
$$+\langle j_0^y(x,y,-z)j_0^y(x',y',z')\rangle+\langle j_0^y(x,y,z)j_0^y(x',y',-z')\rangle]\ . \tag{17}$$

The factor α is chosen to be $1/\sqrt{2}$ so that the integrand in Eq. (17) approaches the bulk value far from the interface. We have checked that our ansatz is consistent with current-conservation requirements such as Eq. (13).

The most general form of conductivity for an isotropic medium is

$$\sigma_{\alpha\beta}(\mathbf{k})=\sigma_\perp(k)\left[\delta_{\alpha\beta}-\frac{k_\alpha k_\beta}{k^2}\right]+\sigma_\parallel(k)\frac{k_\alpha k_\beta}{k^2}\ , \tag{18}$$

where the longitudinal part is irrelevant to our problem. Now we have everything needed for the calculation of the correlation function. Substituting (18) and (10) into (17) we get correct correlation functions which we use in Eq. (9). By performing contour integration in the complex plane we obtain

$$\langle|H^z(z,\omega)|^2\rangle=\frac{2T}{\pi c^2}\int d\mathbf{p}\,\sigma_\perp(p)\frac{q^2}{p^2}e^{-2qz}\ , \tag{19}$$

where $\mathbf{q}=(p_x,p_y)$. In cylindrical coordinates we have

$$\langle|H^z(z,\omega)|^2\rangle$$
$$=\frac{4T}{c^2}\int_0^\infty dp\,\sigma_\perp(p)\int_0^\pi d\theta\sin^3\theta\,e^{-2pz\sin\theta}\ . \tag{20}$$

Now we should choose a model to describe transverse conductivity. We are interested in the static limit ($\omega\rightarrow 0$). The integral in Eq. (20) can be evaluated exactly if, for example, we take $\sigma_\perp(p)=\sigma_0$:

$$\langle|H^z(z,\omega)|^2\rangle=\frac{\pi\sigma_0 T}{c^2 z}\ . \tag{21}$$

Using a Fermi-liquid model with the mean free path l, the conductivity is given by[9]

$$\sigma_\perp(p)=\begin{cases}\sigma_0 & pl\le 3\pi/4\\ \dfrac{3\pi\sigma_0}{4pl} & pl>3\pi/4\end{cases}\ , \tag{22}$$

where $\sigma_0=Ne^2l/p_F$. Then we can evaluate the integral in (20) in two limiting cases:

$$\langle|H^z(z,\omega)|^2\rangle=\frac{\pi\sigma_0 T}{c^2}\frac{1}{z}\ ,\quad z\gg 1\ , \tag{23}$$

$$\langle|H^z(z,\omega)|^2\rangle=\frac{\pi\sigma_0 T}{c^2}\frac{4\ln(l/z)}{l}\ ,\quad z\ll 1\ . \tag{24}$$

The same result can be obtained for $\langle|H^x(z,\omega|^2\rangle$ and $\langle|H^y(z,\omega)|^2\rangle$,with the only difference being a numerical factor:

$$\langle|H^x(z,\omega)|^2\rangle=\langle|H^y(z,\omega)|^2\rangle$$
$$=\tfrac{1}{2}\langle|H^z(z,\omega)|^2\rangle\ . \tag{25}$$

Now we can substitute our results into Eq. (1) to obtain the relaxation rate

$$\frac{1}{T_z}=\frac{2}{3}\frac{1}{T_y}=\frac{2}{3}\frac{1}{T_x}=\frac{\pi\sigma_0 T\gamma^2}{2c^2 l}f(z/l)\ , \tag{26}$$

where T_y and T_z correspond to the magnetic field in the y and z directions (respectively), and $f(r)$ is a dimensionless function given by an integral:

$$f(r)=\frac{4}{\pi}\int_0^\infty dk\,\beta(k)\int_0^\pi d\theta\sin^3\theta e^{-2kr\sin\theta}\ , \tag{27}$$

where β is a dimensionless conductivity:

$$\beta(k)=\begin{cases}1 & k\le 3\pi/4\\ \dfrac{3\pi}{4k} & k>3\pi/4\ .\end{cases} \tag{28}$$

The limiting expressions for $f(r)$ are

$$f(r)=\begin{cases}\dfrac{1}{r} & r\gg 1\\ -4\ln(r) & r\ll 1\ .\end{cases} \tag{29}$$

Actually, the integral over the momentum in (20) should be cut off by Fermi momentum with the result that our calculations are valid for $z\gg 1/k_F$. Also using Eq. (10) for the magnetic-field-correlation function we neglect the existence of a skin effect. This assumption is justified in the limit $\omega\rightarrow 0$, that is, in the case of infinite skin-penetration depth. For the case of finite frequency our results still hold if $z\ll\delta$. To check this we take $\omega=10^9\ \mathrm{s}^{-1}$. In this case we are in the regime of an anomalous skin effect and $\delta=(c^2l/4\pi\omega\sigma)^{1/3}$. Substituting $\sigma=10^{20}\ \mathrm{s}^{-1}$ and $l=10^{-3}$ cm we find $\delta=10^{-4}$ cm which gives the upper bound on z for the validity of our results. It follows that our results for $z\gg l[\langle H(z)^2\rangle\sim 1/z]$ are not applicable for this set of parameters. Proper treatment for this case should involve skin-effect equations.

III. SUMMARY

To compare our results with experiment we notice that hydrogen atoms are adsorbed on the layer of solid hydrogen and liquid helium that covers the copper wall of the cell. The hydrogen atoms may move along the surface with thermal velocity. This motion should not affect the nuclear-spin relaxation rate. To obtain numerical results we assume $T=1$ K, $Ne^2/p_F=1.3\times 10^{23}\ \mathrm{s}^{-1}\ \mathrm{cm}^{-1}$, $\mu=1.4\times 10^{-23}$ erg/G. Substituting numerical values into Eq. (26) we obtain $T_z\approx(4\times 10^4)/f(z/l)$s. The mean free path of the electron in copper at this low temperature is determined by the impurity concentration. We estimate it to be $l = 10^{-3}$ cm for the typical annealed oxygen-free copper commonly used in experiments. The typical thickness of a hydrogen-helium layer is about 3×10^{-6} cm. Thus we are in the $z \ll l$ regime. With these assumptions we obtain the relaxation time of 2000 s that is ten times longer than those reported in Ref. 4. We note also that the data in Ref. 4 show the raise of the relaxation rate with decreasing temperature at $T < 0.4$ K while we predict the linear temperature dependence. We attribute this discrepancy to the following. At low temperature the surface density of adsorbed atomic hydrogen grows with decreasing temperature due to the Boltzmann exponent [the binding energy of H to liquid ^{4}He is 1 K (Ref. 10)]. This makes many-body relaxation processes more important. We believe that special experiments can be designed to look for the predicted relaxation rate and verify the dependence on dielectric wall thickness, mean free path, and temperature.

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APPENDIX

Yet another mechanism similar to the one considered in Ref. 3 may be involved in the explanation of the observed relaxation rate on insulator walls. Let us consider the existence of the nonuniform charge distribution on the surface of the insulator, which gives rise to a random electric field above the surface. It can originate from surface nonregularities, microdefects, etc. This electric field is seen as a magnetic field varying in time in the frame of an adsorbed hydrogen atom moving along the surface. These magnetic-field fluctuations may cause nuclear-spin relaxation. To make a quantative estimate we assume the existence of a spatially varying electric field with the root mean square value $E_0 = 10^3$ V/cm and a length scale parallel to the surface of order $l = 10^{-5}$ cm. The local magnetic field seen by the nuclear spin is $H = (v/c)E$, where $v = 10^4$ cm/s is the average thermal velocity at 1 K. Thus the spatial variation on the length l of the electric field is seen as the temporal variation of the magnetic field at the frequency $\omega = v/l$. We are interested in the spectral density of the mean square value of this varying magnetic field at the frequency $\omega_0 = 10^9$ s^{-1}. We note that in our case ω is of the same order as ω_0. Combining the above relations we have $\langle H(\omega)^2 \rangle = (v^2/c^2) E_0^2 l / v$. From this we obtain a relaxation time of the order of 1000 s. The amplitude of the spatial harmonics of the electric field of the wavelength l decays exponentially in the direction perpendicular to the surface as $e^{-z/l}$ for $z > l$. Thus, in contrast with the fluctuating-magnetic-field mechanism described in the text, the relaxation rate related to this mechanism should decay exponentially as a function of the helium layer thickness.

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